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# “Gleaming, white and deadly”: using lead to track human exposure and geographic origins in the Roman period in Britain

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## Introduction

When one reads of lead, it is rarely long before the Roman world is mentioned. Their two stories are closely intertwined in a tale of necessity, progress, and the “enticing vices” of civilization<sup>1</sup> that has left the world a deadly legacy. It is one we are still mitigating. To engineers, builders and metalworkers, lead possesses irresistible qualities: the metal is relatively inert and resistant to corrosion when in contact with either air or water, has a low melting point (327°C), and is malleable; it is therefore easy to smelt, refine, re-use and work. It is cheap, widely available and can do “a great deal of metallurgical donkey-work”.<sup>2</sup> When its compounds are added to paints, pigments, dyes, petrol, batteries and foodstuffs, they confer qualities hard to replicate by other means. But there is a price. Lead is toxic and deleterious to calcium metabolism and the nervous, digestive and reproductive systems.<sup>3</sup> The clinical symptoms of lead poisoning, such as ‘dry bellyache’, anaemia, nerve disorders, infertility, memory loss and an inability to concentrate, are well-known. In addition, high lead exposure has been associated with malaria, rickets, gout, Paget’s disease, and periodontal disease.<sup>4</sup> Today the toxicity of lead on the neurological development of children, first reported by R. Byers and E. Lord,<sup>5</sup> is blamed for a range of anti-social and delinquent behaviours and compromised intelligence even at sub-clinical levels.<sup>6</sup> The dysfunctional society created by a people poisoned by smelters, piped water, lead cooking vessels, cosmetics and wine sweetened and preserved with *sapa*, *defrutum* and sugar of lead (lead acetate) has been proposed (and just as vehemently refuted) as a contributing factor in the fall of the Roman empire.<sup>7</sup>

In his *Alexipharmaca*,<sup>8</sup> Nicander provides the first known written record (mid-2nd c. B.C.) of “gleaming, deadly white lead whose fresh colour is like milk which foams all over”, along with a graphic description of the symptoms resulting from exposure.<sup>9</sup> Although the Romans knew that exposure to lead fumes and ingestion of lead was injurious to health and spoiled the taste of water,<sup>10</sup> still they continued to mine, smelt, work, use, and ingest it

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1 Tac., *Agr.* 21.

2 Alexander and Street 1989, 187.

3 Clarkson 1987; Goyer 1996.

4 Caffey 1938; Gordon and Whitehead 1949; Nriagu 1983a; Vico and Dessy 1988; Spencer *et al.* 1995; Fullmer 1997; Adachi *et al.* 1998; Saraiva *et al.* 2007; Nriagu *et al.* 2008.

5 Byers and Lord 1943.

6 Needleman *et al.* 2002; Needleman 2009.

7 Gilfillan 1965; Waldron 1973; Hodge 1981; Nriagu 1983b; Curran 1984; Scarborough 1984; Woolley 1984; Needleman and Needleman 1985; Lessler 1988.

8 Nicander, *Alexipharmaca* 2.74 ff.

9 Grout.

10 Vitr. 8.6.1-11, quoted in Humphrey *et al.* 1998, 296-97.

in hitherto-unheard-of quantities. In addition to being present in trace quantities in other metals and alloys, such as silver, copper and bronze, lead metal itself was used to manufacture a wide range of domestic and industrial products, such as water pipes, cisterns, cooking equipment, pewter ware, tokens, slingshot, fishing gear and coffins.<sup>11</sup> Compounds such as lead acetate and lead carbonate were produced for sweeteners, preservatives, medicines, glazes for pottery, pigments and cosmetics.<sup>12</sup> The main, but by no means only, sources of lead were located in Greece, Spain and Britain.<sup>13</sup> Britain was noted to be a particularly rich and easily available source of 'black lead', the silver-bearing lead-rich ore, galena (PbS) which, according to Pliny, "in Britain occurs at the surface of the ground in such abundance that a law forbids the production of more than a certain quantity".<sup>14</sup>

Greek and Roman mining activities are recorded in Greenland ice cores, Icelandic salt marshes, Scandinavian lake sediments and European peat bogs as elevated lead concentrations. These environmental archives all testify to a sharp rise in background atmospheric lead deposition from c.500 B.C., a fall to background levels c.A.D. 500, and then an inexorable rise through the 2nd millennium A.D., finally peaking in the 20th c.<sup>15</sup> Similarly, the lead level in Swedish varve sediments peaks at the beginning of the first millennium A.D., followed by a long 'clean' period between A.D. 400 and 900. This increase in the amount of atmospheric lead is accompanied by a shift in its lead isotope ratios (which vary in different rocks depending on their age and the minerals present) away from that of averaged natural dust (produced by land clearances, soil and rock erosion) towards those of the metal ores being mined.<sup>16</sup> Nonetheless, the amount of lead reaching these ancient environmental archives of atmospheric pollution is infinitesimally small; even today, when a far higher level of atmospheric pollution exists, people in different parts of the world have lead in their bodies that remains indicative of the lead pollution in their country of origin.<sup>17</sup> How then is the lead pollution record reflected in the people who lived with it? Can the use of metal be identified in ancient human populations, or was it only the miners, smelters and smiths who were affected? When does the lead isotope ratio of ancient people stop reflecting the geographic region from which they sourced their food and drink, and start reflecting the culturally-mediated pollution of their environment from metal extraction and use? Since the small but groundbreaking study by T. Molleson *et al.*,<sup>18</sup> very few studies have attempted to use lead isotopes to track the residential mobility of humans in Britain (see Table 11.1).

This paper draws together lead isotope and concentration data obtained for tooth enamel from over 200 archaeological burials at 33 sites in Britain, Ireland and Rome since 1996 (Table 11.1). The aim is to establish if immigrants to Britain during the Romano-British period can be identified on the basis of their differential exposure to pollutant lead. Details

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11 E.g., Boulakia 1972; Toller 1977; Hodge 1981; Beagrie 1989; Boni *et al.* 2000; Rosen and Galili 2007.

12 Humphrey *et al.* 1998; Greene 2007; Van Elslande *et al.* 2008.

13 Alexander and Street 1989, 185.

14 Plin., *NH* 34.164-65, quoted in Humphrey *et al.* 1998, 177.

15 Hong *et al.* 1994; Renberg *et al.* 1994; Shotyk *et al.* 1996; Rosman *et al.* 1997; Weiss *et al.* 1997; De Vleeschouwer *et al.* 2007; Marshall *et al.* 2009.

16 Brännvall *et al.* 1999.

17 Gulson *et al.* 1997.

18 Molleson *et al.* 1986.

TABLE 1

THE SITES FROM WHICH ARCHAEOLOGICAL ENAMEL SAMPLES WERE TAKEN FOR USE HERE, with references for the data. Note that the 4 samples from Poundbury were bone.

<i>Period</i>	<i>Location</i>	<i>Site name</i>	<i>Source of data</i>
Neolithic	Shetland	Sumburgh	this publication
	Dorset	Monkton-up-Wimbourne	Montgomery 2000
Neolithic/Early Bronze Age	Yorkshire	West Heslerton	Montgomery <i>et al.</i> 2005
Bronze Age	Yorkshire	Callis Wold	this publication
	Yorkshire	Towthorpe	this publication
	Yorkshire	Aldro	this publication
	South Uist	Cladh Hallan	this publication/Parker Pearson <i>et al.</i> 2005
	Mull	Ardachy	this publication
	Lewis	Cnip	this publication
	Yorkshire	Gristhorpe	this publication
Iron Age	Yorkshire	Wetwang	this publication
	Yorkshire	West Heslerton	Montgomery <i>et al.</i> 2005
	Rome	Castellaccio Europarco	this publication
Scottish/Irish Iron Age	Dublin	Rath	this publication
	Dublin	Ratoath	this publication
	Isle of Skye	High Pasture Cave	this publication
	Orkney	Mine Howe	this publication
	Lewis	Galson	this publication
	South Uist	Kilpheder	this publication
	York	3/6 Driffield Terrace	Montgomery <i>et al.</i> forthcoming/this publication
1st-4th c. A.D.	Winchester	Eagle Hotel	Montgomery 2002
	Bristol	Mangotsfield	Montgomery 2002
	London	Spitalfields	Montgomery 2002
	Dorset	Poundbury	Molleson <i>et al.</i> 1986
	Rome	Casal Bertone	this publication
	Rome	Castellaccio Europarco	this publication
	Yorkshire	West Heslerton	Montgomery <i>et al.</i> 2005
	Warwickshire	Wasperton	this publication
	Yorkshire	Riccal	Budd <i>et al.</i> 2004
	Derbyshire	Repton	Budd <i>et al.</i> 2004
8th-11th c. A.D.	Orkney	Westness	this publication
	Dublin	Great St George St.	this publication
	Lewis	Cnip	this publication
	Cumbria	St Bees	Knusel <i>et al.</i> 2010
	Gloucester	Blackfriars	Montgomery 2002
	Orkney	Graemsay	this publication
	Hereford	Hereford Cathedral	this publication

of the preparation and measurement are in the Technical Appendix. Initial results suggest a non-British origin for a very wealthy female burial from Spitalfields in London, and for 4 males excavated from a decapitation cemetery at Driffield Terrace at York.

## The effect of large-scale lead mining on ambient lead ratios and human exposure in Britain

A lead-bead necklace found in an Early Bronze Age cist burial from the Scottish Borders is currently the earliest evidence for the use of lead metal in Britain.<sup>19</sup> Isotope analysis of the tiny beads has demonstrated that the lead is consistent with sources in the Borders region at Wanlockhead/Leadhills.<sup>20</sup> Although this does not preclude the beads being imported from a congruent foreign source, the find suggests that the technology to mine and smelt lead, perhaps as a source of silver, may have existed long before large-scale lead mining commenced in Britain.<sup>21</sup> The lead ore deposits in Britain occur predominantly in the West and North, hosted mainly in Carboniferous limestone sequences. Figure 11.1 shows the locations of the lead ore deposits believed to have been exploited during the Romano-British period on the basis of historical records or finds of lead ingots (pigs) bearing Latin inscriptions.<sup>22</sup> The earliest Romano-British lead pig from Wookey Hole (Somerset) is dated to A.D. 49;<sup>23</sup> together with others dating to the reign of Vespasian (A.D. 69-79), it suggests that large-scale mining was underway in the Mendips, Flintshire and Derbyshire by this time,<sup>24</sup> both as a by-product of silver extraction and, in the case of the virtually silver-free Derbyshire ore, in its own right.<sup>25</sup> Given the ease with which lead can be recycled, it is likely that it is under-represented in the Romano-British archaeological record.<sup>26</sup> It was, however, being used for piping water by A.D. 79, as attested by examples from Chester which are stamped with the name of the governor, Gnaeus Iulius Agricola.<sup>27</sup> The richness of the British ore deposits enabled lead to be exported. One route was via Gaul: the wreck of the *Ploumanac'h* contained 271 lead ingots with inscriptions referring, perhaps understandably, to the Brigantes who inhabited the lead-rich Pennine uplands and, less obviously, the Iceni in the southeast where no lead deposits occur.<sup>28</sup> Britain was still regarded as a significant source of lead during the

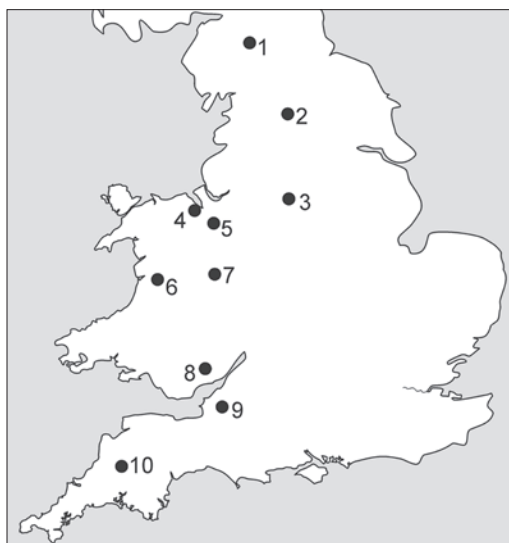


Fig. 11.1. Location of the British lead ore sources exploited during the Romano-British period: 1. Alston and the Pennines; 2. Yorkshire Dales; 3. Derbyshire; 4. Halkyn Mt.; 5. Fridd; 6. Plynlimon; 7. Linley; 8. Draethen; 9. Charterhouse; 10. Cornwall (redrawn from Jones and Mattingly 2002).

19 Hunter and Davis 1993.

20 Hunter 2000.

21 Tylecote 1987, 41.

22 E.g., Whittick 1931; 1982; Collingwood and Myers 1937; Elkington 1976; Jones and Mattingly 2002.

23 *RIB* II.1, 2404.1.

24 *RIB* II.1, pp. 38-61.

25 Tylecote 1992, 71.

26 M. McCarthy, pers. comm.

27 *RIB* II.3, 2434.1-2 and 2434.3.

28 Frere *et al.* *RIB* II.1, 66.

Early Mediaeval period.<sup>29</sup> Possibly a considerable amount of Romano-British lead metal remained in circulation and was recycled during the Early Mediaeval period, with smelting of new ore taking place on a far smaller scale than in the Romano-British period.<sup>30</sup> This would explain the reduction in atmospheric lead deposition attested in the environmental archives.

The first study to seek evidence for lead poisoning in ancient people by measuring the amount present in their excavated bones was carried out by S. Jarcho<sup>31</sup> on a population in the American Southwest known to produce lead-glazed pottery, but in this case the levels of lead were low (below 20 mg kg<sup>-1</sup>) and not thought to be indicative of toxic exposure. Subsequently, proof of the high levels of skeletal lead in inhabitants of the Roman world that would be indicative of lead poisoning has been searched for and apparently found, with reported bone lead levels falling in the region of 300 mg kg<sup>-1</sup>.<sup>32</sup> By comparison, typical modern adult bone lead concentrations are ~3–60 mg kg<sup>-1</sup>.<sup>33</sup> In juveniles, research has indicated that bone lead levels of just 25 mg kg<sup>-1</sup> are a proxy indicator for blood lead levels that are detrimental to neurological development, and thus a good predictor of criminal and delinquent behaviour.<sup>34</sup> However, measuring lead in archaeological bone or dentine is problematic, particularly given the known ability of bone meal to ‘mop up’ the lead present in the contaminated soils to which it is applied.<sup>35</sup> Lead accumulation during burial has been demonstrated for both dentine and bone.<sup>36</sup> The amount of lead incorporated into the bone during burial is difficult to establish but it will depend on the local conditions (e.g., soil pH, temperature and hydrology). If the isotope ratios are measured, it may be possible to demonstrate that those of the bone are *different* to those of the mobile lead in the soil, but proving that the bone is *entirely* free of diagenetic lead is problematic.<sup>37</sup> Notwithstanding this, some studies have reported very low levels of lead in the bones of prehistoric or unpolluted populations compared with those measured from the Roman, mediaeval and modern periods.<sup>38</sup>

Alongside the uncertainties associated with measuring trace elements in bone that has been buried come the uncertainties of attempting to identify lead poisoning (as opposed to lead exposure) by measuring lead concentrations in bone samples from individuals of different ages. These arise because skeletal lead (1) has a long residence time (>27 years) in the skeleton; (2) is subject to individual health and metabolic status; and (3) can accumulate with age and can be released back into the blood stream many years after it was deposited.<sup>39</sup> Moreover, bone is a living, evolving tissue and is subject to modelling (formation)

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29 Bayley 1992, 6.

30 Ibid. 6.

31 Jarcho 1964.

32 Mackie *et al.* 1975; Drasch 1982; Jaworowski *et al.* 1985; Molleson *et al.* 1986; Grandjean 1988; Aufderheide *et al.* 1992.

33 Gross *et al.* 1975; Drasch 1982; Jaworowski *et al.* 1985; Manea-Krichthen *et al.* 1991; Arnay-De-La-Rosa *et al.* 1998; Yoshinaga *et al.* 1998.

34 Needleman *et al.* 2002.

35 Ma *et al.* 1993; Lower *et al.* 1998; Hodson *et al.* 2001.

36 Waldron 1981, 1982 and 1983; Whittaker and Stack 1984; Lambert *et al.* 1985; Ericson 1993; Budd *et al.* 1998; Montgomery 2002; Chiaradia *et al.* 2003; Zapata *et al.* 2006.

37 Montgomery 2002; Bower *et al.* 2005; De Muynck *et al.* 2008.

38 Drasch 1982; Jaworowski *et al.* 1985; Corruccini *et al.* 1987; Ericson *et al.* 1991; Arnay-De-La-Rosa *et al.* 1998; Yoshinaga *et al.* 1998; Wittmers *et al.* 2002.

39 Rabinowitz *et al.* 1973; Gross *et al.* 1975; Barry 1978; Gulson *et al.* 1998; Gulson *et al.* 1999.

and remodelling (turnover) processes throughout a person's lifetime. These are functionally different and proceed at different rates at different times of life in the skeleton. As a consequence, lead is not distributed homogeneously throughout the skeleton and concentrations can vary substantially between different bones.<sup>40</sup> It is thus not easy to prove that the amount of lead measured in a single bone sample is representative of the whole skeleton, if it is indicative of a single period of acute high-level exposure at some distant period of life, or if it has amassed as a result of life-long low-level exposure that may not have produced any clinical symptoms or deficiencies. Hair would represent a better archive of short-term or recent exposure, but it is rarely available at archaeological sites and *post-mortem* contamination is still a major consideration. Using enamel as a shorter-term 'snapshot' of lead exposure removes many of the age and health related intra- and inter-tissue variables and greatly reduces the risk of *post-mortem* contamination.<sup>41</sup> It does, however, limit the time of life being investigated to childhood, and specifically to the years when the enamel of that tooth was forming. It is possible that a child will not be exposed to some of the high-lead sources an adult may encounter (e.g., wine and *sapa*), so that enamel will under-estimate adult exposure. However, until their gut is fully developed, children are far more susceptible to lead poisoning than adults because the latter are able to excrete most of the lead they ingest.<sup>42</sup> Moreover, lead is known to cross the mammalian mammary and placental barriers, and tissue lead levels are very similar between mother and child.<sup>43</sup> Indeed, it was noted over a century ago that foetal and infant mortality were very high amongst female lead workers, and that the women used pregnancy as a way of removing lead from their own bodies, knowing that the infant would not survive.<sup>44</sup>

### Enamel lead concentration data from Britain

Figure 11.2 is compiled from data in Tables 11.2 and 11.3 and sources in Table 11.1. It illustrates how childhood lead exposure changes from the Neolithic to the Late Mediaeval period in Britain. Rarely do concentrations rise above 0.4 mg kg<sup>-1</sup> until the 1st c. A.D., and in places such as Shetland and Yorkshire they are routinely below 0.2 mg kg<sup>-1</sup>. The only individuals from the Iron Age who have lead concentrations of 1 mg kg<sup>-1</sup> or higher are from N Britain and from the Long Iron Age (i.e., contemporary with the Romano-British and Early Mediaeval period of southern Britain). The data define a log-normal distribution; the median enamel lead concentration for prehistoric individuals in Britain is 0.07 mg kg<sup>-1</sup> (n=54). There is little comparative enamel data from 'unpolluted' modern populations, but this is, for example, an order of magnitude lower than the mean of 0.33 mg kg<sup>-1</sup> found in the enamel of a remote, unpolluted rural population from S Africa.<sup>45</sup> The blood lead level required to produce an enamel lead concentration of 0.07 mg kg<sup>-1</sup> is estimated to be 0.7 µg dL<sup>-1</sup> (i.e. a ratio of µg dL<sup>-1</sup> blood : mg kg<sup>-1</sup> enamel of ~10 : 1).<sup>46</sup> This is well below the current World Health Organisation's advisory blood lead limit for children of 10 µg dL<sup>-1</sup>,

40 Aufderheide 1989; Erkkilä *et al.* 1992.

41 Ericson 1993; Montgomery 2002.

42 Bowen 1979, 125; Mahaffey 1978.

43 Mahaffey 1991; Franklin *et al.* 1997.

44 Oliver 1914; Smith 1986.

45 Grobler *et al.* 2000.

46 *Ibid.*; Arora *et al.* 2006.



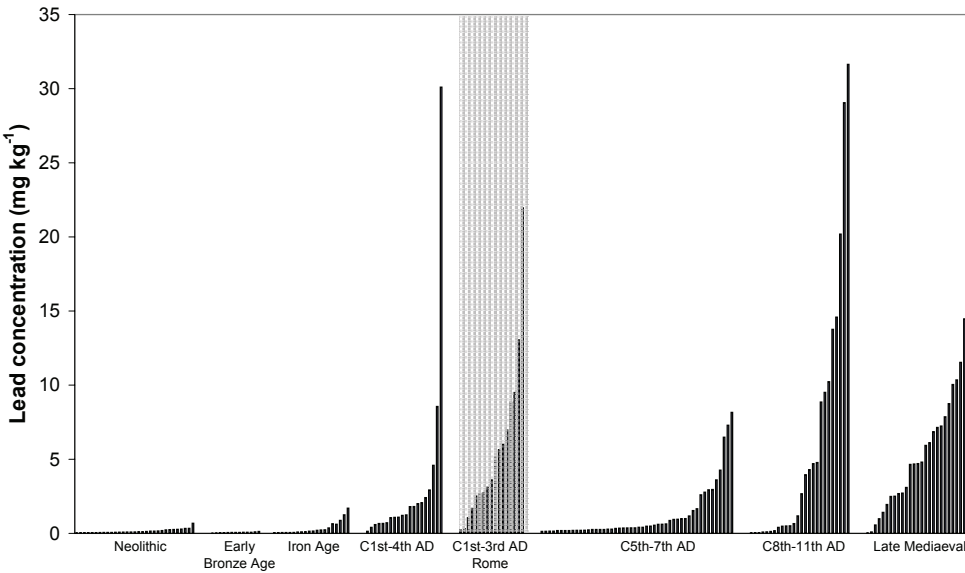


Fig. 11.2. Enamel lead concentrations ( $\text{mg kg}^{-1}$ ) of burials from Britain dating from the Neolithic to the Late Mediaeval period. Samples are plotted in order of increasing concentrations within each time period. Comparative data for Rome of the 1st to 3rd c. A.D. are highlighted in grey. The data comes from Tables 11.2-3 and sources listed in Table 11.1.

TABLE 11.2  
ENAMEL LEAD CONCENTRATIONS ( $\text{MG KG}^{-1}$ )  
OBTAINED BY QUADRUPOLE INDUCTIVELY COUPLED PLASMA MASS SPECTROMETRY (ICP-MS)  
at the British Geological Survey. See Technical Appendix for details of measurement and errors.

	Location	Site name	Sample no.	Pb $\text{mg kg}^{-1}$
Neolithic	Shetland	Sumburgh	Sumb-2	0.04
			Sumb-5	0.06
			Sumb-6	0.03
			Sumb-9	0.06
			Sumb-11	0.05
			Sumb-12	0.08
			Sumb-15	0.13
			Sumb-16	0.09
	Yorkshire	Callis Wold	CW-12	0.19
			CW-13	0.16
		Towthorpe	TP-17	0.10
			TP-16	0.07
		Aldro	AL-19	0.27
			AL-20	0.15
			AL-21	0.09
			AL-23	0.11
			AL-22	0.11
Bronze Age	Mull	Ardachy	MULL-1	0.06
			BIPSK-92	0.07
			BIPSK-122	0.13
	Yorkshire Wolds	Acklam Wold	BIPSK-125	0.07
			CGSU-105	0.10
	South Uist	Cladh Hallan	CHSU-5087	0.06
			CHSU-5424	0.06
			CHSU-5424	0.06
Iron Age	Yorkshire	Wetwang	WWH-14	0.04
			WWH-121	0.14
			WWH-161	0.06



	<i>Location</i>	<i>Site name</i>	<i>Sample no.</i>	<i>Pb mg kg<sup>-1</sup></i>
1st-2nd c. A.D.	Rome	Castellaccio Europarco	WWH-173	0.09
			WWH-275	0.06
			WWH-348	0.05
			WWH-351	0.15
			WWH-388	0.06
			ET58	0.24
			ET27	0.32
			ET72	2.52
			ET67	2.68
			ET42	3.61
			ET31	6.97
			ET69	9.50
		Casal Bertone	CBT70	1.06
			CBT18	1.68
			CBT36	2.73
			CBT23	3.11
			CBT30	5.17
			CBF12A	5.64
			CBF1A	6.00
			CBF11A	8.85
			CBT33	13.0
			CBT32	21.9
Early Mediaeval	Warwickshire	Wasperton	Wasp-1	4.26
			Wasp-6	1.54
			Wasp-27	0.99
			Wasp-28	0.63
			Wasp-35	0.61
			Wasp-42	0.37
			Wasp-46	0.95
			Wasp-48	2.94
			Wasp-55	2.60
			Wasp-153	6.49
			Wasp-174	2.78
			Wasp-180	3.61
			Wasp-190	0.36
			Wasp-191	0.61
			Wasp-193	7.29
			Wasp-194	1.17
			Wasp-195	0.33
Late Mediaeval	Hereford	Hereford Cathedral	H140	14.5
			H410	7.15
			H438	2.51
			H713	4.71
			H741	2.68
			H869	4.80
			H905	7.86
			H911	0.56
			H959	6.11
			H1517	2.72
			H1774	3.10
			H1798	0.98
			H2006	2.49
			H221	10.3
			H2374	1.42
			H2656	4.68
			H3265	5.93
			H341	1.94
			H3661	4.65
			H3850	11.5

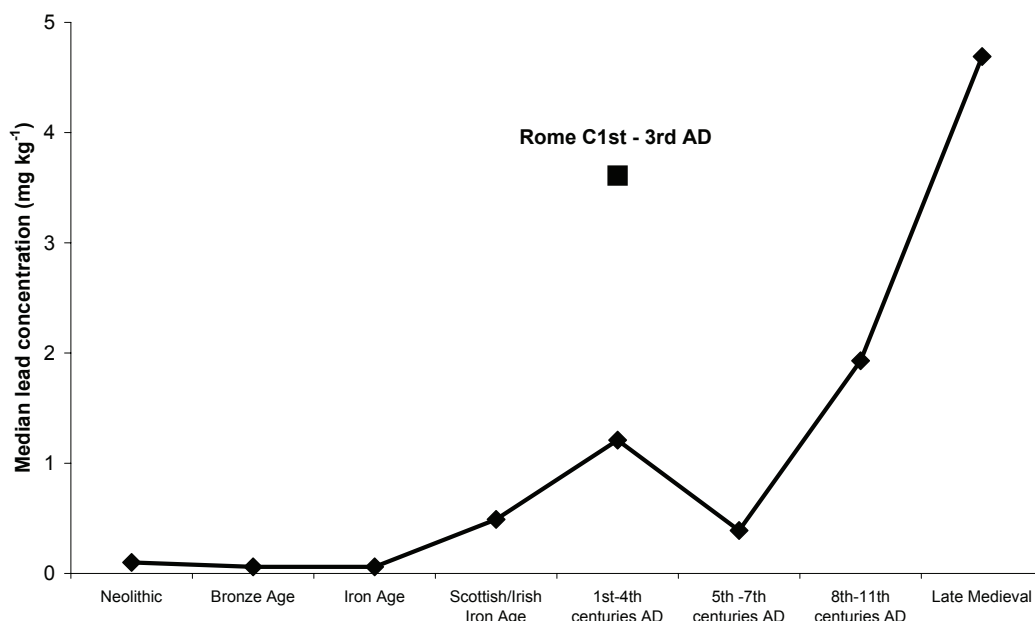


Fig. 11.3. Median human enamel lead concentrations in Britain (see Table 11.4), from the Neolithic to the Late Mediaeval period, illustrating diachronic trends in exposure to anthropogenic lead. The median human lead concentration for Rome of the 1st to 3rd c. A.D. is included for comparison.

although current research is suggesting this limit should be reduced to  $5 \mu\text{g dL}^{-1}$ .<sup>47</sup> According to the above blood : enamel ratio, a 'safe' blood lead limit of  $5 \mu\text{g dL}^{-1}$  would produce enamel lead below  $0.5 \text{ mg kg}^{-1}$  which is in good agreement with the prehistoric human data presented in fig. 11.2: all but one are below  $0.5 \text{ mg kg}^{-1}$ .

From the 1st c. A.D., some individuals exhibit enamel lead concentrations of up to  $30 \text{ mg kg}^{-1}$ . These individuals have a level of lead that is 10,000 times higher than that of the least polluted individual in this study: the Early Bronze Age skeleton from Gristhorpe (Yorkshire) ( $3 \mu\text{g kg}^{-1}$ ; Table 11.3). According to the ratio above, an enamel lead concentration of  $30 \text{ mg kg}^{-1}$  would arise from a blood lead level of  $c.300 \mu\text{g dL}^{-1}$ , which is far higher than the  $\sim 100 \mu\text{g dL}^{-1}$  associated with "very severe poisonings".<sup>48</sup> The median enamel lead burden for individuals living in Rome during the first few centuries A.D. is higher than that for Britain ( $3.6 \text{ mg kg}^{-1}$ , compared to  $1.2 \text{ mg kg}^{-1}$ ) (fig. 11.3), but the wide range of concentrations are comparable (Table 11.4). However, it should be noted that the Italian data is derived from two cemeteries only (Casal Bertone and Castellaccio Europarco) which probably housed only lower-class individuals.<sup>49</sup> It is therefore possible that the lead concentrations observed are not fully representative of all groups of Imperial society.

There is little difference in the median lead levels of Neolithic people and those in the Bronze and Iron Ages to suggest that simply living in a mining or metal-using society increases lead exposure in children (fig. 11.3). This observation would be supported by modern studies of children inhabiting highly-polluted lead mining villages, such as Broken Hill (Australia) and Winster (Derbyshire), where body lead burdens were found to be

<sup>47</sup> Binns *et al.* 2007.

<sup>48</sup> Levin and Goldberg 2000.

<sup>49</sup> Buccellato *et al.* 2008; Musco *et al.* 2008.

TABLE 11.3  
ENAMEL LEAD ISOTOPE RATIOS AND ELEMENTAL CONCENTRATION  
OBTAINED BY THERMAL IONISATION MASS SPECTROMETRY (TIMS)  
at the NERC Isotope Geosciences Laboratory, British Geological Survey.  
Some samples (denoted by \*) were measured by multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS).  
See Technical Appendix for details of measurement and errors.

<i>Period</i>	<i>Location</i>	<i>Site</i>	<i>Sample no.</i>	<i>Pb mg kg<sup>-1</sup></i>	<i><sup>206</sup>Pb/<sup>204</sup>Pb</i>	<i><sup>207</sup>Pb/<sup>204</sup>Pb</i>	<i><sup>208</sup>Pb/<sup>204</sup>Pb</i>	<i><sup>207</sup>Pb/<sup>206</sup>Pb</i>	<i><sup>208</sup>Pb/<sup>206</sup>Pb</i>
Bronze Age	Yorkshire	Gristhorpe	GRIS-1	0.003	18.45	15.63	38.44	0.8471	2.0830
			Cnip BA	0.06	17.93	15.49	38.32	0.8641	2.1376
Scottish Iron Age	Lewis, Outer Hebrides	Cnip	RATO-1	0.65	18.85	15.64	38.45	0.8296	2.0398
			RATH-848	0.36	18.68	15.60	38.29	0.8352	2.0502
	Dublin	Rath	MH05 1861	0.25	18.32	15.62	38.31	0.8525	2.0917
	Dublin	Mine Howe	MH04-897	0.31	18.35	15.63	38.42	0.8517	2.0939
	Orkney		HPCS-100	-	18.3454	15.6137	38.3670	0.85112	2.09137
Isle of Skye	High Pasture Cave		HPCS-101	-	18.3477	15.6148	38.3823	0.85109	2.09194
			HPCS-102	0.87	18.3633	15.6285	38.4337	0.85108	2.09297
			HPCS-103	-	18.3572	15.6244	38.4228	0.85119	2.09307
			Gal-93	1.25	18.45	15.62	38.42	0.8464	2.0826
			Gal-96	0.61	18.50	15.74	39.35	0.8525	2.1276
Lewis, Outer Hebrides	Galson		Gal-II	0.23	18.16	15.59	38.35	0.8584	2.1116
			Gal-IV	1.70	18.42	15.63	38.40	0.8485	2.0845
			Gal-V	0.10	18.15	15.58	38.62	0.8585	2.1277
			SUK-1	0.05	18.27	15.66	38.45	0.8575	2.1045
			SUK-2	0.05	18.08	15.60	38.14	0.8627	2.1095
4th-3rd c. B.C.	Rome	Castellaccio Euro-parco	ET93	3.27	18.4467	15.6525	38.5978	0.84854	2.09239

<i>Period</i>	<i>Location</i>	<i>Site</i>	<i>Sample no.</i>	<i>Pb mg kg<sup>-1</sup></i>	<i><sup>206</sup>Pb/<sup>204</sup>Pb</i>	<i><sup>207</sup>Pb/<sup>204</sup>Pb</i>	<i><sup>208</sup>Pb/<sup>204</sup>Pb</i>	<i><sup>207</sup>Pb/<sup>206</sup>Pb</i>	<i><sup>208</sup>Pb/<sup>206</sup>Pb</i>
1st-4th c. A.D.	York	Driffield Terrace	Drif-18	1.06	18.4433	15.6346	38.4232	0.84755	2.08331 *
			6Drif-9	1.24	18.5977	15.6249	38.5068	0.84002	2.07051 *
			Drif-15	1.21	18.4249	15.6394	38.4266	0.84891	2.08557 *
			Drif-21	2.00	18.4643	15.6344	38.4568	0.84676	2.08276 *
			lead coffin	-	18.4001	15.6291	38.3686	0.84935	2.08524 *
London	Rome	Spitalfields Castellaccio Euro- parco	ET31	6.97	18.4012	15.6577	38.5597	0.85090	2.09550 *
			CBF11A	8.85	18.5316	15.6611	38.6948	0.84511	2.08804 *
			WSTN-5	0.13	18.46	15.60	38.39	0.8456	2.0801
			WSTN-12	0.03	18.41	15.44	38.29	0.8384	2.0793
			WSTN-28A	0.18	18.30	15.55	38.15	0.8489	2.0845
Dublin	Great St George St.		WSTN-25	0.09	18.61	15.52	38.64	0.8344	2.0765
			WSTN-32	0.10	18.56	15.55	38.51	0.8382	2.0750
			F598-2T	0.27	18.57	15.59	38.32	0.8392	2.0633
			F196-3T	0.29	18.46	15.63	38.33	0.8468	2.0767
			F223-1T	0.25	18.51	15.58	38.35	0.8418	2.0717
Lewis, Outer Hebrides	Cnip		A	1.18	18.39	15.58	38.31	0.8473	2.0830
			B	-	18.52	15.60	38.45	0.8424	2.0755
			F	0.66	18.36	15.56	38.35	0.8474	2.0886
			D	0.48	18.43	15.59	38.33	0.8455	2.0790
			D	0.50	18.40	15.59	38.26	0.8471	2.0799
Late Mediaeval	Orkney	Graemsay	E	-	18.60	15.65	38.41	0.8414	2.0652
			C	-	18.45	15.61	38.40	0.8458	2.0813
			G	-	18.48	15.60	38.42	0.8445	2.0792
			GRAE-1	0.02	17.62	15.46	37.58	0.8777	2.1330

TABLE 11.4

MEDIAN ENAMEL LEAD CONCENTRATIONS (MG KG<sup>-1</sup>) BY PERIOD

<i>Period</i>	<i>Median (mg kg<sup>-1</sup>)</i>	<i>Range (mg kg<sup>-1</sup>)</i>	<i>n</i>
Neolithic	0.10	0.03-0.68	31
Bronze Age	0.06	0.003-0.13	13
Iron Age	0.06	0.04-0.15	10
All Prehistoric	0.07	0.003-0.68	54
Scottish/Irish Iron Age	0.49	0.10-0.65	10
1st-4th c. A.D.	1.21	0.24-30.1	25
1st-3rd c. A.D. Rome	3.61	0.24-21.9	17
5th-7th c. A.D.	0.39	0.13-8.16	50
8th-11th c. A.D.	1.93	0.03-31.6	26
Late Mediaeval	4.69	0.02-14.5	26

much lower than expected and within the normal range for urban-dwelling children. This is because the lead from the mine-waste that polluted the environment was highly insoluble pyromorphite (a lead chloride phosphate) and therefore not bioavailable.<sup>50</sup> Conversely, people living within two miles of lead smelters have significantly raised blood lead levels, both from inhaling fumes and living in an environment which is polluted with very small lead particles, both of which facilitate lead uptake in the gut.<sup>51</sup> In lead-using societies, the principal causes of lead poisoning in polluted humans are the following: drinking soft water or acidic fluids from lead pipes, cisterns and vessels (the calcium in hard water reduces the uptake of lead by the gut to negligible quantities); deliberate ingestion of bioavailable lead compounds, such as lead acetate and lead carbonate; and accidental ingestion of lead compounds by the use of lead or lead-glazed pottery, pans, storage containers, cosmetics and medicines.<sup>52</sup>

Throughout the last two millennia, individual lead exposure has been highly variable: there are still many exhibiting very low levels of lead, particularly in the 5th to 11th c. A.D. when the median lead concentration drops to 0.39 mg kg<sup>-1</sup> (n=50). This coincides with the 'clean' period in the archives of atmospheric deposition mentioned above; it suggests that the most hazardous sources of lead exposure, such as the inhalation of smelting fumes or ingestion of bioavailable lead compounds in contaminated food, drink and medicines, were reduced during the Early Mediaeval period. From the 8th c. A.D., human lead burdens rise in line with the environmental archives mentioned above. Although not illustrated here, the rise in childhood lead exposure continues through to the 19th c., when remarkably high levels approaching 100 mg kg<sup>-1</sup> have been recorded in adults who nonetheless managed to survive that level of childhood exposure.<sup>53</sup> In modern populations, enamel lead concentrations in excess of 10 mg kg<sup>-1</sup> are rare;<sup>54</sup> from their studies at the Broken Hill lead mining village, B. Gulson and D. Wilson<sup>55</sup> concluded that high exposure produced enamel lead concentrations of ~2-10 mg kg<sup>-1</sup>.

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50 Cotter-Howells and Thornton 1991; Gulson *et al.* 1996.

51 Mahaffey 1978.

52 Ibid.

53 Trickett 2008.

54 Losee *et al.* 1974; Farmer *et al.* 1994; Gulson *et al.* 1997; Montgomery 2002.

55 Gulson and Wilson 1994; Gulson 1996.

### How did Neolithic people obtain their body lead burden?

Lead is naturally present, albeit in very small quantities, in most of the Earth's rocks, soils and waters,<sup>56</sup> whence it can be taken up by plants or adhered to their surfaces before being ingested by animals. It is a non-nutrient trace element that is ingested and metabolised into mammalian tissues principally from food and drink, although inhalation can be an important pathway: adults absorb only 5-10% via the digestive system, but retain 30-50% of inhaled lead from atmospheric pollution.<sup>57</sup> As already mentioned, infants and children are particularly susceptible to lead poisoning: they absorb lead far more efficiently across the gut wall than do adults;<sup>58</sup> lead is more readily taken up by animals fed an entirely liquid diet, which implies the same may be true for human infants;<sup>59</sup> and soil is a significant source of lead both through soil pica and general hand-to-mouth activity.<sup>60</sup> The ingestion of soil and dust via poorly washed hands, food and vessels, dirty water and grit in flour would have been possible throughout prehistory, but differences in the prevalence of such practices, as well as geographical variation resulting from different rocks and differential access to hard or soft water sources, mean that regional differences in the magnitude of natural human lead burdens would still exist. Further work is needed to characterize such variation.

The vast majority of the lead in a human body is found in the skeleton, where it is commonly believed to occupy calcium sites within the bioapatite lattice,<sup>61</sup> although there is scant hard evidence in the literature to support a simple substitution for calcium in any mineral because the two elements are geochemically dissimilar;<sup>62</sup> moreover, the two elements are antagonistic and the route of lead to the skeleton is different to that of calcium (lead, like zinc, is primarily transported in the red blood cells, calcium, like strontium, in the plasma).<sup>63</sup> As W. and M. Neuman stated over 50 years ago, "its entry into the crystal does not involve a simple displacement of calcium as heretofore believed ... This 'bone-seeker' needs reinvestigation".<sup>64</sup> It is therefore to be expected that trace levels of lead would be present in the skeletons of people living in pre-metallurgical societies. In such societies, the isotope ratio of the skeletal lead will be indicative of the rock from which it originated and, like strontium isotopes, can be used to provide information about geographic origins.<sup>65</sup> For example, Cretaceous Chalk has a  $^{207}\text{Pb}/^{206}\text{Pb}$  ratio of ~0.830, and recent marine carbonate shell sand, such as machair, is even lower, at ~0.813;<sup>66</sup> older silicate rocks can provide much higher  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios. The cost and the increased complexity of interpreting several isotope ratios (as opposed to the one of strontium) is probably the reason lead has not been used more widely in this manner. However, there is another factor. Unlike strontium, the natural skeletal lead isotope ratios obtained from country rocks can be swamped if large amounts of lead from ore sources with different isotope profiles are mined, smelted and

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<sup>56</sup> Faure 1986.

<sup>57</sup> Underwood 1977, 413.

<sup>58</sup> Bowen 1979.

<sup>59</sup> WHO 1996, 198.

<sup>60</sup> Mahaffey 1978; Jaworowski *et al.* 1985; Mielke and Reagan 1998.

<sup>61</sup> Verbeeck *et al.* 1981; Wallach and Chausmer 1990, 240.

<sup>62</sup> Faure 1986.

<sup>63</sup> Bowen 1979, 106.

<sup>64</sup> Neuman and Neuman 1958, 94-95.

<sup>65</sup> Gulson 1986; Molleson *et al.* 1986; Montgomery *et al.* 2000.

<sup>66</sup> Montgomery 2002.

introduced into the environment. If such lead is transported into regions from which it does not originate (as much of it is), the link between a person and the rocks from which they sourced their food will be severed. In southern Britain, this appears to happen in the first centuries A.D., when most individuals exhibit very similar lead isotope ratios that bear no relation to their particular region of origin but derive from ambient pollutant lead consistent with (although not unique to) ore sources from southern Britain.<sup>67</sup>

Figure 11.4 shows the relationship between the amount of lead present in enamel and one of its isotope ratios, in this case  $^{207}\text{Pb}/^{206}\text{Pb}$ . Two observations can be made: first, all prehistoric individuals have uniformly low lead concentrations but highly variable ratios; second, lead concentrations in excess of  $\sim 0.5 \text{ mg kg}^{-1}$  (i.e., above the dotted line) generally occur when the lead has an isotope ratio between 0.845 and 0.849. A few individuals, all dating to the Romano-British period, do not conform to this trend and will be discussed in more detail below. The observed range of ratios is consistent with ore lead from mines in southern Britain.<sup>68</sup> This diachronic human exposure-pattern mirrors that seen in the environmental archives.<sup>69</sup> It results from an increase in mining and smelting activity and the use of a wide variety of manufactured products made of, or containing, lead. This phenomenon has been termed “cultural focussing”<sup>70</sup> — that is, as anthropogenic pollution increases, the population’s lead burden increases and their isotope ratios converge on a point within the ore lead field. In human terms, fig. 11.4 illustrates that enamel lead concentrations above  $0.5 \text{ mg kg}^{-1}$  require some contribution from pollutant lead, and those of  $1 \text{ mg kg}^{-1}$  are derived predominantly from pollutant lead that has been mined and smelted; in southern Britain, such individuals will have  $^{207}\text{Pb}/^{206}\text{Pb}$  between 0.845 and 0.849. Individuals from 19th-c. England also conform to this trend<sup>71</sup> — until, that is, the increasing import of Australian lead shifts the environmental ratios towards higher  $^{207}\text{Pb}/^{206}\text{Pb}$  values by the start of the 20th c.<sup>72</sup> Thus, the isotope ratio of an individual with  $1 \text{ mg kg}^{-1}$  or more of lead will be wholly or predominantly derived from pollutant sources, and the ratios will reflect the average lead pollution an individual was exposed to and able to access in their cultural sphere, rather than the geological terrain where they source their food and drink. Whilst this homogenisation of the lead isotope ratios renders them ineffectual at tracking the movement of most people *within* southern Britain during the last 2000 years, it should make those exposed to pollutant lead from foreign sources (with a different lead isotope signature) relatively easy to identify. Moreover, given the considerable variability in cultural lead exposure of people from similar time periods, the combination of lead concentration and ratios might, for example, allow us to discriminate rural from urban origins, high-status from low-status, or a ‘civilized’ from a ‘non-civilized’ society.

### Cultural focussing of the lead isotope ratios

Figure 11.4 uses one lead isotope ratio ( $^{207}\text{Pb}/^{206}\text{Pb}$ ), but the 4 lead isotopes that can be measured ( $^{204}\text{Pb}$ ,  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$  and  $^{208}\text{Pb}$ ) can be combined in various ways. Unlike strontium, which is conventionally presented as  $^{87}\text{Sr}/^{86}\text{Sr}$  in most scientific literature, different

67 Ibid.

68 Barreiro 1995; Haggerty *et al.* 1996; Rohl 1996; Shepherd *et al.* 2009.

69 Brännvall *et al.* 1999.

70 Montgomery *et al.* 2005.

71 Kowal *et al.* 1991; Trickett 2008.

72 Bacon *et al.* 1996.



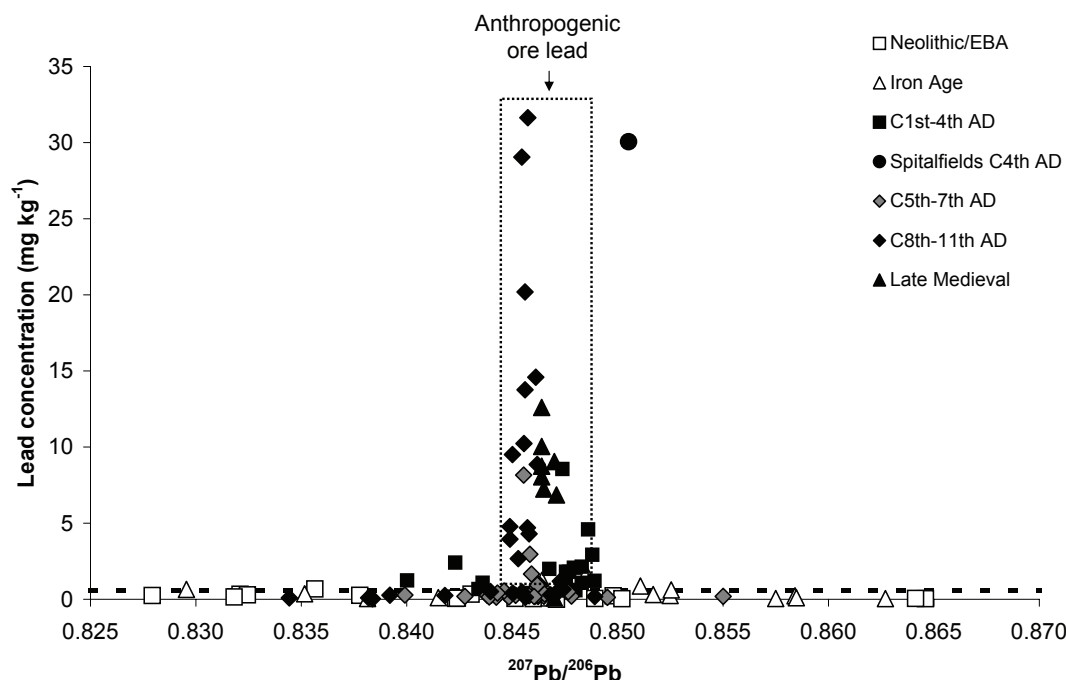


Fig. 11.4. A plot of human enamel lead concentrations and  $^{207}\text{Pb}/^{206}\text{Pb}$  isotope ratios. The horizontal dashed line indicates the maximum level of lead ( $0.5 \text{ mg kg}^{-1}$ ) that the data suggests can be obtained by unpolluted individuals. Note that most individuals above this line have very similar  $^{207}\text{Pb}/^{206}\text{Pb}$  isotope ratios, consistent with lead ores in southern Britain. The few individuals outside this field (indicated by the dotted rectangle) are discussed in the text. The data come from Table 11.3 and sources listed in Table 11.1. 2s errors for TIMS and MC-ICP-MS data are within the symbols.

disciplines tend to favour particular combinations of lead isotopes and plot different ratios against each other. By ratioing  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$  and  $^{208}\text{Pb}$  to  $^{204}\text{Pb}$ , isotope data can be viewed in three dimensions in which related samples may cluster or define a linear array, but this is difficult to do clearly on the printed page. Publications will often present only combinations, or even just one (if that one discriminates sufficiently between the samples), of the three radiogenic isotopes  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$  and  $^{208}\text{Pb}$  — e.g.,  $^{208}\text{Pb}/^{206}\text{Pb}$ ,  $^{206}\text{Pb}/^{207}\text{Pb}$  or  $^{207}\text{Pb}/^{206}\text{Pb}$  — because non-radiogenic  $^{204}\text{Pb}$  is more difficult to measure due to its low abundance and reduced precision.

In figs. 11.5 and 11.6, the human enamel data is plotted by period (as defined in Table 11.1) and lead concentration, respectively. Figure 11.5 shows how only prehistoric individuals have highly variable lead isotope ratios which derive solely or predominantly from natural lead exposure from country rock sources. Above and to the left of the main field, which is inside the box, are individuals excavated from the Northern and Western Isles of Scotland: these reflect the ancient Scottish lead ores and Pre-Cambrian basement rocks.<sup>73</sup> Those below and to the right were excavated from English and Irish marine carbonate rocks. The majority of individuals from the Romano-British period onwards cluster tightly inside the box on the same trend as, and in the same region occupied by, galena from English and Welsh mines.<sup>74</sup>

<sup>73</sup> Dickin 1981.

<sup>74</sup> Rohl 1996; Barreiro 1995; Haggerty *et al.* 1996.

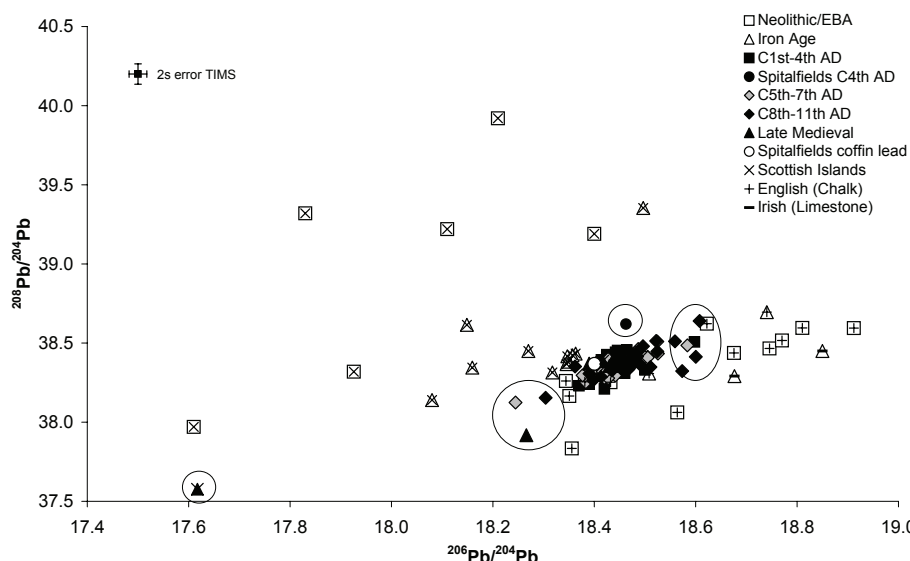


Fig. 11.5. A plot of  $^{208}\text{Pb}/^{204}\text{Pb}$  versus  $^{206}\text{Pb}/^{204}\text{Pb}$  for human enamel lead, dating from the Neolithic to the Late Mediaeval period, grouped by period. Note that most prehistoric individuals (i.e., with little exposure to anthropogenic lead) are loosely grouped according to their regional origin by the combination of their lead isotope ratios. All the individuals above and to the left of the main field were excavated from the Northern and Western Isles of Scotland (x), while those below and to the right were excavated from marine carbonates in England (+) and Ireland (-). Most individuals from the 1st to 16th c. A.D. cluster tightly in a diagonal array, illustrating their 'culturally focussed' lead isotope ratios. The remaining individuals (circled), who plot outside the main cluster despite dating from the 5th c. A.D. onwards, are discussed in the text. 2s errors for MC-ICP-MS data are within the symbols.

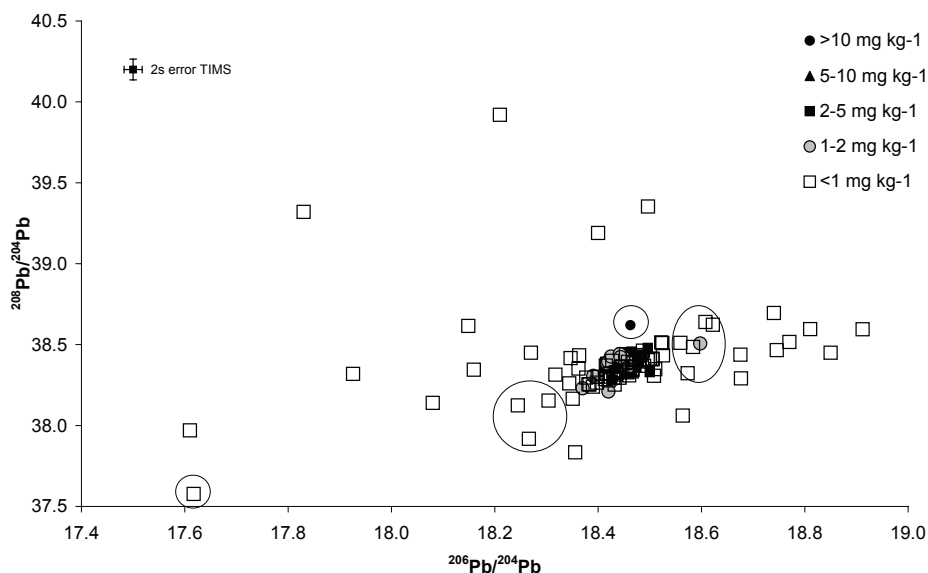


Fig. 11.6. A plot of  $^{208}\text{Pb}/^{204}\text{Pb}$  versus  $^{206}\text{Pb}/^{204}\text{Pb}$  for human enamel lead, dating from the Neolithic to the Late Mediaeval period. Individuals are grouped based upon the level of lead present in their enamel. The circled individuals are the same as those circled in fig. 11.5: this figure shows that, for the majority, their position outside the main cluster is explained by the low levels of lead. 2s errors for MC-ICP-MS data are within the symbols.

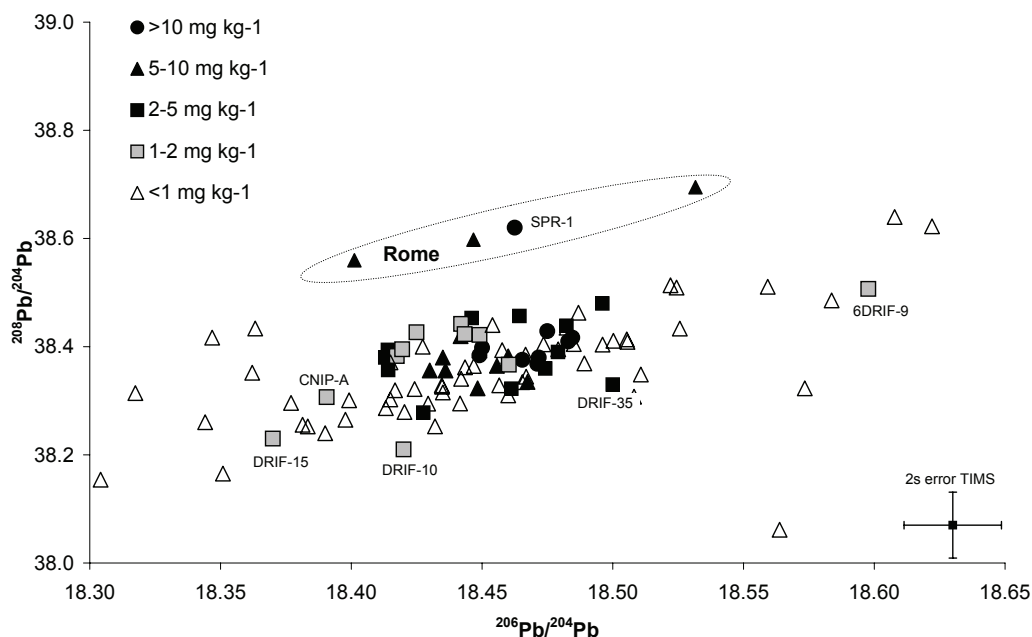


Fig. 11.7. A plot of  $^{208}\text{Pb}/^{204}\text{Pb}$  versus  $^{206}\text{Pb}/^{204}\text{Pb}$  for human enamel lead, dating from the Neolithic to the Late Mediaeval period, magnified from fig. 6. Individuals are plotted according to the amount of lead present in their enamel. The dashed oval highlights 3 samples from Rome. 2s errors for MC-ICP-MS data (e.g., York Driffield Terrace and Rome) are within the symbols.

There are 4 groups of individuals (circled) who do not conform to this trend; for the majority this is because, despite dating from the 5th c. A.D. onwards, they exhibit a very low level of lead (fig. 11.6), which suggests a childhood with little or no exposure to pollutant sources. Their isotope ratios will thus be partly or wholly derived from the region where they sourced their food and drink. Only two polluted individuals fall outside the central field of humans exposed to English ore lead and both are of Romano-British date, one being the young adult female burial from Spitalfields in London (black circle; SPR-1; fig. 11.6) and the other an adult male burial from 6 Driffield Terrace in York (grey circle; 6DRIF-9). The clustering seen in other archaeological humans dating from the last two millennia strongly suggests that these two individuals would have been unlikely to obtain such a lead isotope signature whilst in southern Britain, unless during childhood they consumed primarily imported foodstuffs or food prepared in imported vessels. In view of the ready availability of British lead in this period, this scenario seems unlikely, but it is a possibility that cannot be entirely ruled out.

The lead isotope range of individuals raised in southern Britain and exposed to indigenous pollutant lead can perhaps be constrained even further. Figure 11.7 contains a magnified region from fig. 11.6, showing the central polluted group in greater detail. The polluted individuals (i.e., those with 1 mg kg<sup>-1</sup> or more of lead) who do not cluster in the central group are generally of Romano-British date. There is one exception: CNIP-A, a female buried in the 8th-10th c. on the Isle of Lewis, Outer Hebrides, with a grave assemblage typical of Viking burials.<sup>75</sup> She does not appear to be of Hebridean origin and there

75 Welander *et al.* 1987.

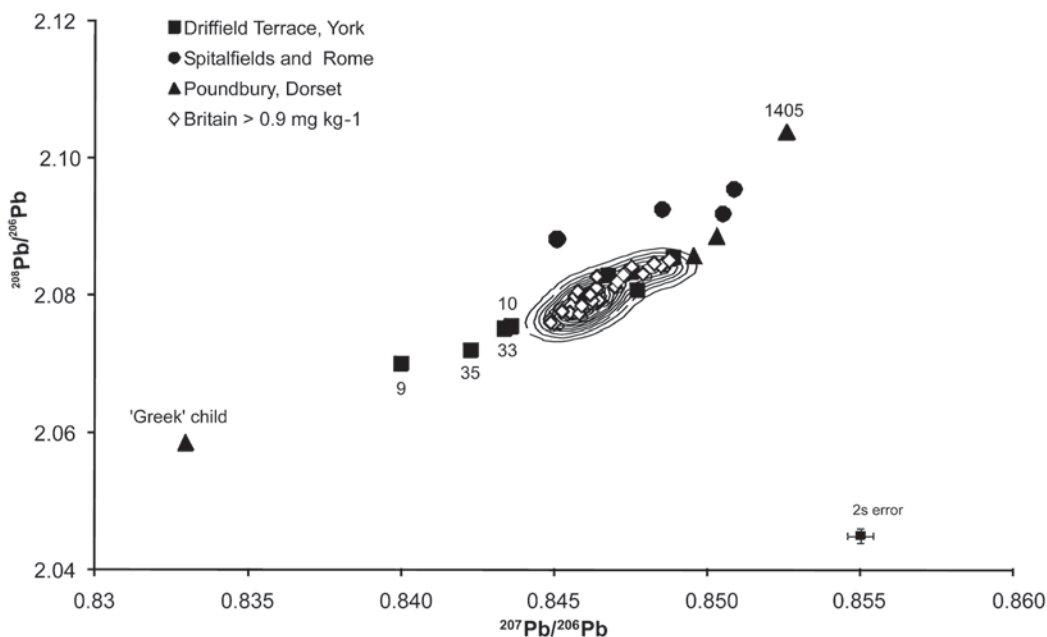


Fig. 11.8. A plot of  $^{208}\text{Pb}/^{206}\text{Pb}$  versus  $^{207}\text{Pb}/^{206}\text{Pb}$ , showing only polluted British burials (i.e., those with  $> 0.9 \text{ mg kg}^{-1}$  of lead in their enamel). The contours for the field of polluted individuals are produced from 35 samples by bivariate Kernel Density Estimate (KDE) analysis using GCDkit application (Janouek *et al.* 2006). Bone data from 4 individuals in the cemetery at Poundbury (Dorchester) are also included (Molleson *et al.* 1986). The density plot is in agreement with the 0.845-0.849 range indicated by the 'ore lead' rectangle in fig. 4. Individuals from York Driffild Terrace, London Spitalfields, Rome, and Poundbury fall outside this oval, which indicates that their lead comes from a different source. Younger Mesozoic ore deposits plot to the bottom left and older Cambrian ores to the top right. 2s errors for MC-ICP-MS data (e.g., Driffild Terrace York and Rome) are within the symbols.

is no reason to believe she originates from southern Britain.<sup>76</sup> The remainder are the female burial of the 4th c. A.D. from Spitalfields, London (SPR-1), three individuals from Rome, and 4 males from the cemetery of the 2nd-4th c. A.D. at Driffild Terrace, York (6DRIF-9, DRIF-10, DRIF-15 and DRIF-35). It is possible that these latter individuals could define a linear array below the main British field, but some could equally be within error of it. Lead isotope fields can overlap and intersect in three-dimensional space, and a single bi-plot may not be sufficient to demonstrate a clear difference. This is a consideration for characterising fields and assigning samples to them. In this instance, a plot of  $^{208}\text{Pb}/^{206}\text{Pb}$  versus  $^{207}\text{Pb}/^{206}\text{Pb}$  (fig. 11.8) can more clearly illustrate the relationship between York's Driffild Terrace individuals and other polluted individuals from southern Britain.

### Can we use anthropogenic lead exposure to identify immigrants to Britain?

Figure 11.8 compares data for polluted humans excavated from southern Britain with three groups of burials dating to the Romano-British period. The contours for the field of polluted individuals are produced from 35 samples by bivariate Kernel Density Estimate (KDE) analysis using GCDkit application.<sup>77</sup> On this plot, younger ores of Mesozoic-Tertiary date plot towards the bottom left, whilst older ores of Cambrian and Hercynian date plot

<sup>76</sup> Montgomery and Evans 2006.

<sup>77</sup> Janouek *et al.* 2006.

to the upper right; those of southern Britain, being predominantly of Carboniferous date, fall in the centre.<sup>78</sup> The kernel density estimate for the polluted individuals shows that the human lead field falls within the ore field for southern Britain, but the distribution of human data is more restricted and does not reflect the discrete geographical clusters of ores present in major ore fields such as the Mendip.<sup>79</sup> The field for polluted humans defines a single cluster with a focus at ~0.846 (and thus in agreement with the peak in fig. 11.4), suggesting that humans are negating, through averaging and mixing processes, the influence of ore field outliers and the distinct clustering of individual ore veins.

Three individuals interred at Rome define a linear array above the field for polluted humans (figs. 11.7-8); this group also encompasses the female burial from London Spitalfields and is discussed in detail below. Four individuals from York Driffeld Terrace (DRIF-10, DRIF-33, DRIF-35 and 6DRIF-9) define a linear array to the left of the polluted human field. Younger ore sources, such as the Tertiary deposits of the Mediterranean basin and the Near East, can provide such lead isotope ratios.<sup>80</sup> On this basis, it is unlikely that these 4 individuals from this unusual, predominantly male cemetery, where many individuals appear to have been executed by decapitation,<sup>81</sup> had origins in Rome or southern Britain. This conclusion is supported by evidence for diet and origins obtained from other isotope systems.<sup>82</sup>

### The female burial from Spitalfields, London

The young adult female excavated in 1999 by the Museum of London Archaeology Service at the Roman burial ground at Spitalfields was sealed in a lead coffin decorated with an intricate scallop-shell pattern and contained within a limestone sarcophagus (fig. 11.9). The coffin was excavated at a depth of 6 m below the modern ground surface. Fine glass and jet grave goods were recovered along with the remains of silk woven with gold thread and bay leaves. A date in the early 4th c. A.D. has been established for the burial.<sup>83</sup> Obtaining lead isotope data from an individual who has spent almost 1700 years in a lead coffin is somewhat problematic. However, the core enamel sample was prepared from the second premolar with the utmost care<sup>84</sup> and, although the enamel lead concentration of 30 mg kg<sup>-1</sup> is high, the isotope ratios bear no resemblance to those obtained from the coffin, which indicate the coffin was probably made of English lead (fig. 11.5). An enamel lead level of 30 mg kg<sup>-1</sup> suggests sufficient lead was circulating in the body while the enamel was forming to cause significant clinical symptoms of lead poisoning. The presence on the tooth of a black gingival lead line,<sup>85</sup> a symptom of adult lead poisoning,<sup>86</sup> suggests that a high-lead intake persisted into adulthood. The enamel lead isotope ratio is unusual for British burials (being enriched in <sup>208</sup>Pb and <sup>207</sup>Pb) and at the time no sources could be identified. Similarly, an adult male (1405) buried with hobnails at his feet and within a wooden coffin from the

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78 Boni *et al.* 2000.

79 Haggerty *et al.* 1996.

80 Stos-Gale *et al.* 1996; Ponting *et al.* 2003.

81 Hunter-Mann 2005; Ottaway 2005.

82 Montgomery *et al.* forthcoming; Eckardt *et al.* above in this volume; Müldner *et al.* forthcoming.

83 Thomas 1999.

84 Montgomery 2002, 165.

85 Ibid.

86 Levin and Goldberg 2000.



Fig. 11.9. The young woman buried in the decorated lead coffin within a limestone sarcophagus at London Spitalfields (© MOLAS).

mainly 4th-c. cemetery at Poundbury near Dorchester<sup>87</sup> was also found to have enriched ratios (fig. 11.8). Here too the authors were unable to identify any possible sources for the lead, although they did not believe that imported food and drink were the cause.<sup>88</sup> Lead with isotope ratios that indicated a younger ore source, such as those of Laurion (Greece), were obtained from the bones of a young child, and the authors concluded it was of Greek origin (fig. 11.8). However, as we now know, such ratios are also consistent with the Cretaceous Chalk in which it was interred,<sup>89</sup> and, because the lead was measured in bone rather than enamel, it is difficult to know if the lead was of *ante* or *post-mortem* origin. Further analyses of the tooth enamel would be useful in this case.

The strontium isotope ratio of 0.7099 obtained from the Spitalfields (London) burial<sup>90</sup> is not particularly diagnostic because it is consistent with origins on Mesozoic sediments which occur widely in SE England<sup>91</sup> and across large areas of N and S Europe. It did not, therefore,

help to clarify the situation, leading us to conclude at the time:

clearly, in the absence of data from contemporary inhabitants of Rome, it is impossible to know if such a combination of strontium and lead would be compatible with an origin there or elsewhere in the Roman Empire.<sup>92</sup>

Very recently, just such data have become available. Included in figs. 11.7-8 are lead isotope data from three individuals at Castellaccio Europarco (ET31 and ET93) and Casal Bertone (CBF11A), two archaeological populations curated by the anthropology division of the Soprintendenza Archeologica di Roma.<sup>93</sup> The site of Casal Bertone, located c.1.5 km east of the city walls just off the *via Praenestina*, was an urban area that has produced remains of a nymphaeum, a burial area consisting of an above-ground mausoleum and a necropolis, and a large industrial complex that was either a fullery or a tannery.<sup>94</sup> Skeletons from this site are assumed to be those of lower- to middle-class individuals, based on the simple

87 Farwell and Molleson 1993.

88 Molleson *et al.* 1986.

89 Montgomery 2002.

90 Ibid.

91 Evans *et al.* 2010.

92 Montgomery 2002, 170.

93 See Killgrove above in this volume.

94 Musco *et al.* 2008.



burial style and lack of grave goods. At Castellaccio Europarco, burials took place in the *suburbium* along the *via Laurentina*, c.12 km south of the city; they are probably associated with a nearby villa. The site was used as a burial ground in three different periods: 4th–3rd c. B.C., 2nd–1st c. B.C., and 1st–2nd c. A.D.<sup>95</sup> All burials were simple in nature and the interred individuals are thought to have been of the lower classes. Full publication of the skeletal remains from these two sites is forthcoming.<sup>96</sup>

These three samples from Rome define a linear array above the field for southern Britain and the array encompasses SPR-1 (figs. 11.7–8). Although no single corresponding ore source can currently be identified, such an array could arise as a result of exposure to lead from the multitude of sources circulating and mixing in Rome.<sup>97</sup> These individuals, for example, are consistent with the lead isotope field characterised for silver *denarii* struck at Rome mints during Severus' eighth imperatorship.<sup>98</sup> This connection may at first sight seem tenuous, but it is quite possible that a ready local market existed for the lead from which this silver was cupelled for coinage. Further evidence to add weight to the argument for a single origin for these 4 individuals comes from the strontium isotope ratios, which are virtually identical.<sup>99</sup> Taken together, and on the basis that the lead isotope ratios rule out Britain as a source of origin for the young female buried at London Spitalfields, the data provide a strong case for a childhood origin in Rome for this wealthy lady.

## Conclusions

As more enamel lead isotope data is obtained from British burials, those who are different can be identified with increasing confidence. In southern Britain, the cultural focussing of human enamel lead isotope ratios as a result of widespread mining, smelting, mixing and use of lead-bearing compounds and artefacts since the end of the Iron Age has resulted in the vast majority of indigenous polluted people (i.e., those with 1 mg kg<sup>-1</sup> or more lead in tooth enamel) having very similar lead isotope ratios (<sup>207</sup>Pb/<sup>206</sup>Pb of 0.845 to 0.849). These ratios appear to be averaged values of ambient pollution produced by the main lead-bearing ores of Carboniferous date which in themselves have a far wider range of ratios. Prehistoric individuals and those with very little cultural exposure to pollutant lead (i.e., less than 0.5 mg kg<sup>-1</sup> in tooth enamel) have far more variable isotope ratios that reflect the rocks in their region of origin. The appearance of increased lead concentrations only during the Romano-British period supports the observation that people “had little use for (lead) until the plumbing revolution of Greco-Roman times”,<sup>100</sup> and it was the cultural use of lead in the food supply to transport, contain, adulterate, preserve and prepare it that is at the root of increased human lead exposure, not whether a society possessed the technology to obtain and use metal.

This cultural focussing of the lead isotope ratio enables lead-polluted individuals from the Romano-British period to be identified as immigrants to Britain if their lead isotope ratios do not conform to the field identified for lead-polluted humans in Britain. It is

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95 Buccellato 2007; Buccellato *et al.* 2008.

96 Killgrove 2010.

97 Boni *et al.* 2000.

98 Ponting *et al.* 2003.

99 Killgrove 2010.

100 Tylecote 1987, 41.



particularly useful in situations when other isotope systems, such as strontium, cannot discriminate between individuals from regions of Mesozoic rocks which occur widely across southern Britain and Europe: Britons do not have a lead isotope ratio that derives from Mesozoic lead ores. This is the case for 4 male burials from the decapitation cemetery in York at Driffild Terrace who appear to have ingested lead from Mesozoic-Tertiary sources, such as those of the Mediterranean basin: their lead ratios are inconsistent with Britain and, on current evidence, Imperial Rome.

A high-status female burial from Spitalfields in London was also found to have lead and strontium isotope ratios that are consistent with three individuals from Rome. Despite being buried in a lead coffin, the isotope ratios of the tooth enamel were very different from that of the coffin, which appears to have been made of British lead. This is the first Roman burial from Britain for which strong corroborating evidence indicates an origin in Rome itself.

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### TECHNICAL APPENDIX

Lead isotope analysis was conventionally undertaken using a Thermal Ionisation Mass Spectrometer (TIMS). However, in recent years the newer technique of Multi Collector Inductively Coupled Plasma Mass Spectrometer (known as MC-ICP-MS, or sometimes PIMMS) has been used. This has the advantage of reducing the analytical error on the lead analysis by as much as an order of magnitude because of the use of a Thallium internal fractionation monitor. Data from both analytical techniques are included in this paper. The paper reviews data produced over the last 10 years, and the vast majority were obtained using the older TIMS method.<sup>101</sup> We provide a general methodology for both techniques while referring the reader to the original publications, where available, for details of the specific study.

Error bars on the diagrams are given for the TIMS technique, but readers should be aware that those samples marked with an asterisk in Table 11.3 have significantly better reproducibility, having been analysed by the MC-ICP-MS method.

#### *Sample preparation*

In a clean laboratory, the enamel samples were first cleaned ultrasonically in high purity water to remove dust, rinsed twice, dried in high purity acetone, and then weighed into pre-cleaned Teflon beakers. The tooth sample was dissolved in Teflon distilled 16M HNO<sub>3</sub> and then an aliquot of c.20% was pipetted into another cleaned Teflon beaker. For determining the lead concentration by TIMS analysis, a 20% aliquot was spiked with <sup>208</sup>Pb-tracer solution for lead concentration analysis, using

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<sup>101</sup> Details of the methods can be found in the following references: Weeks *et al.* 2009, Montgomery *et al.* 2005.

the isotope dilution method. Lead was then collected from all the samples using conventional anion exchange methods.<sup>102</sup>

#### *TIMS analysis*

The lead isotope composition and concentrations were determined by Thermal Ionisation Mass spectroscopy (TIMS) using a Finnigan Mat 262 multi-collector mass spectrometer. The reproducibility of the NBS982 lead standard may vary with time, but the values used for this study are those given as:<sup>103</sup>

$$^{206}\text{Pb}/^{204}\text{Pb} \pm 0.1\%, \quad ^{207}\text{Pb}/^{204}\text{Pb} \pm 0.13\%, \quad ^{208}\text{Pb}/^{204}\text{Pb} \pm 0.16\%, \quad ^{207}\text{Pb}/^{206}\text{Pb} \pm 0.05\% \text{ and } ^{208}\text{Pb}/^{206}\text{Pb} \pm 0.05\%. \text{ (2s).}$$

#### *MC-ICP-MS analysis*

Lead isotope analysis of the samples was conducted using one of three plasma machines available at NERC Isotope Geosciences Laboratory: VG P54; Nu Plasma HR; or a VG Axiom MC-ICP-MS. Prior to analysis, the samples were centrifuged at 13,000 rpm for 10 minutes and then spiked with a Thallium solution (added to allow for the correction of instrument-induced mass bias). The samples were then diluted with 2% HNO<sub>3</sub>, (i.e., to generate an ion beam intensity less than the Faraday collector saturation point of 10V) and introduced into the instrument via an ESI 50 µl/min PFA micro-concentric nebuliser, attached to a desolvating unit (either a Nu Instruments DSN 100 or Cetac Aridus, depending on the mass spectrometer being used). Five ratios were simultaneously measured ( $\text{Pb}^{206}/\text{Pb}^{204}$ ,  $\text{Pb}^{207}/\text{Pb}^{204}$ ,  $\text{Pb}^{208}/\text{Pb}^{204}$ ,  $\text{Pb}^{207}/\text{Pb}^{206}$  and  $\text{Pb}^{208}/\text{Pb}^{206}$ ). Each acquisition consisted of 75 sets of ratios, collected at 5-second integrations.

The precision and accuracy of the method were assessed through the analysis of an NBS 981 lead standard solution (also spiked with Thallium), run at regular intervals throughout the analytical session. The average values obtained for each of the measured NBS 981 ratios were then compared to the known values for this standard.<sup>104</sup> The sample data was subsequently normalised, according to the relative daily deviation of the measured standard value from the true. Normalisation to an international standard in this way effectively cancels out the effects of slight daily variations in instrumental accuracy, and allows the direct comparison of the data obtained during different analytical sessions. The analytical errors reported for each of the sample ratios are also propagated relative to the respective reproducibility of this standard, to take into account the errors associated with the normalisation process. Typical propagated errors for this technique are taken from a recent unpublished study:

$$^{206}\text{Pb}/^{204}\text{Pb} \pm 0.012\%, \quad ^{207}\text{Pb}/^{204}\text{Pb} \pm 0.012\%, \quad ^{208}\text{Pb}/^{204}\text{Pb} \pm 0.018\%, \quad ^{207}\text{Pb}/^{206}\text{Pb} \pm 0.004\% \text{ and } ^{208}\text{Pb}/^{206}\text{Pb} \pm 0.008\%. \text{ (2s)}$$

#### *Elemental analysis*

Some samples only have concentration data and these were run using a quadrupole ICP-MS. The elemental concentrations of the enamel samples and quality control materials were determined using an Agilent 7500cx quadrupole ICP-MS instrument. The instrument was calibrated using a series of synthetic chemical solutions diluted from multi-element stock solutions (SPEX Certprep<sup>TM</sup>), the calibration being validated using synthetic chemical standards from a separate source. The digest solutions were diluted such that the calcium concentration was between 100 and 200 ppm, optimal for long-term instrument stability, best detection limits, and all elements falling within the defined calibration range. The reproducibility of the lead concentration data is  $\pm 10\%$  (2s).

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